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Laser Damage of Dichroic Coatings in a High Average Power Laser Vacuum Resonator

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Laser damage of dichroic coatings in a high average power laser vacuum resonator

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ABSTRACT

In our application, dichroics in a high average power, near-infrared, laser system have short operating lifetimes. These dichroics were used as the resonator fold mirrors and permited the transmission of the pumping argon (Ar) ion laser light. Representative samples of two different dichroic optics were taken off-line and the transmission performance monitored in various scenarios. Irradiating these optics under resonator vacuum conditions, ($\leq$ 1 mT, 11.7 kW/cm^2, Ar laser running all wavelengths) resulted in a degradation of transmission with time. Irradiating these optics in a rarefied oxygen atmosphere (1 to 10 T of oxygen, 11.7 kW/cm^2, Ar laser running all wavelengths) the transmission remained steady over a period of days. The transmission loss observed in the optic tested in vacuum was somewhat reversible if the optic was subsequently irradiated in a rarefied oxygen atmosphere. This reversibility was only possible if the transmission degradation was not too severe. Further tests demonstrated that an atmosphere of 10 T of air also prevented the transmission degradation. In addition, tests were performed to demonstrate that the optic damage was not caused by the ultra-violet component in the Ar ion laser. Mechanisms that may account for this behavior are proposed.

1. INTRODUCTION

Atomic vapor laser isotope separation (AVLIS) was the largest scale demonstration of high average power lasers. The Lawrence Livermore National Laboratory operated an AVLIS Laser Demonstration Facility that used many different types of high power lasers to enrich uranium.[1] In support of the enrichment activities, a near-infrared (nIR) laser system was constructed of six 25-watt Ar ion lasers pumping with all Ar lines into three Ti:Al_2O_3 rods within a resonator cavity (Figure 1). [2] The cavity supported lasing in a vacuum in order to minimize air turbulence. The optic of interest was the fold mirror located near the ends of the laser rods. The fold mirror was a dichroic coating which was designed to pass Ar wavelengths into and reflect the nIR wavelength within the resonator cavity. One of the dichroic requirements was to pass 1 kW/cm^2 of Ar ion laser light and another to reflect 55 kW/cm^2 intracavity of a nIR wavelength. The coating specifications was typical of others that the project required to survive high laser power densities in a continuous wave (cw) or quasi-cw operating mode. However upon operation, the nIR laser output immediately degraded and fell below operating specifications within hours under the given fluence conditions. [3]

The purpose of this paper is to report results from dichroics tested in an off-line set-up. The set-up was intended to replicate optical degradation, then characterize the effect, and potentially assist in optimizing the dichroic coating design and deposition process.

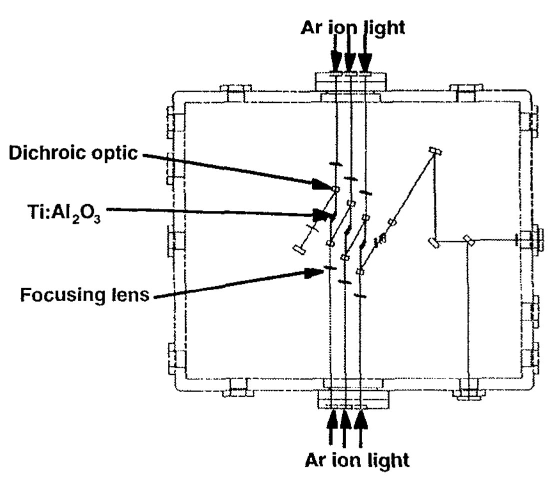


Figure 1 Resonator cavity sketch.

2. TEST SET-UP

Given the same coating specifications, dichroics of two designs (referred to as 900L and AAA) were created independently, made by the same coating supplier, and tested in transmission. Figure 2 shows the transmittance of typical coatings made from each design. The materials selected for each coating design were coincidentally the same. Vacuum tests without laser illumination were done with an EG&G goniometer. As expected, coatings of both designs shifted shorter in spectral performance but showed no transmission degradation after residing 14 days in < 1 mTorr of vacuum.

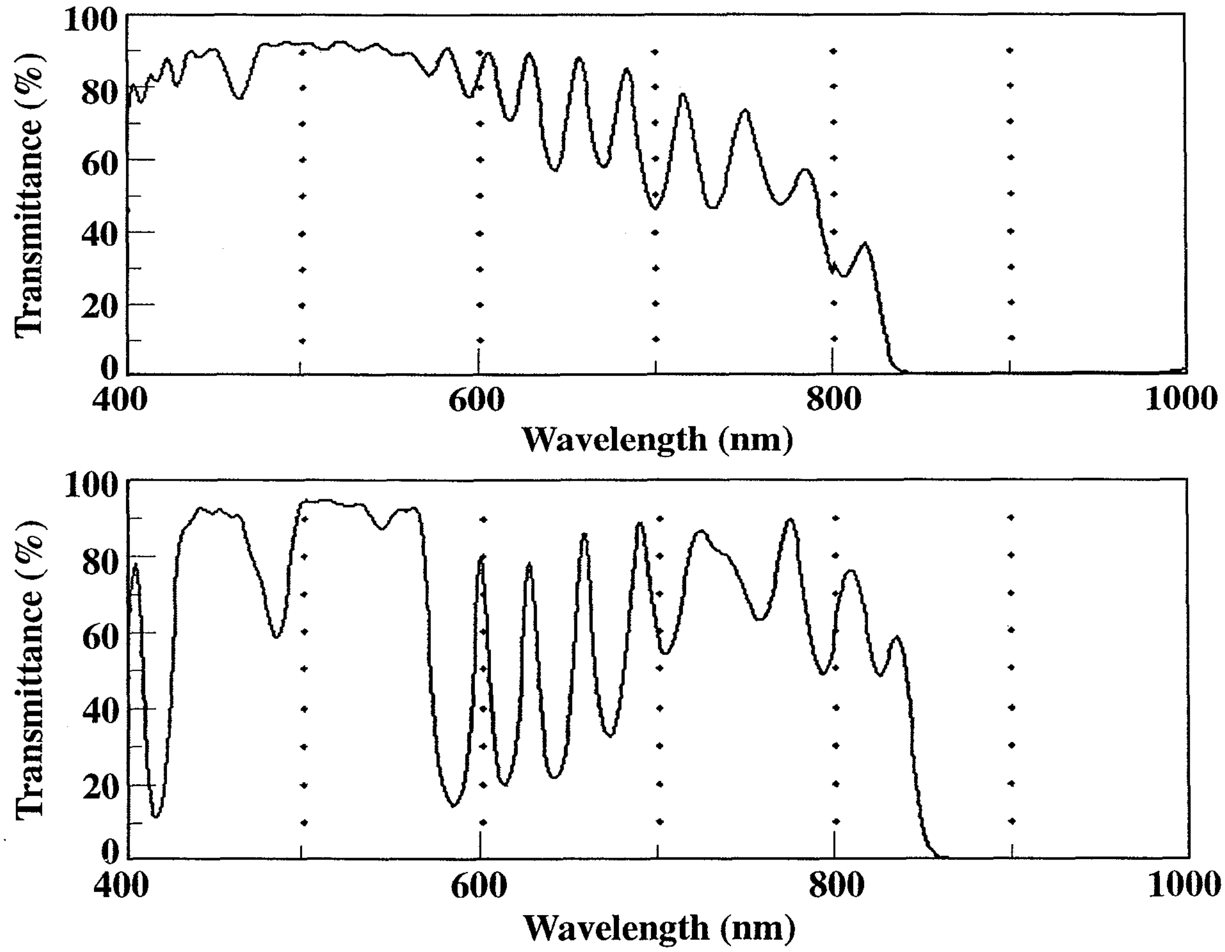


Figure 2 Spectral performance of the dichroic coatings. The upper scan is from a 900L designed coating. The lower scan is from an AAA designed coating.

A six-way stainless steel cross was fitted with a variable angle-of-incidence holder for the dichroic optics (Figure 3). The dichroics were tested at 15° angle-of-incidence. A Coherent Innova-100 laser irradiated the sample with 8 to 15 watts of power at the wavelengths listed in Table 1. The power density was achieved by focusing through a lens to obtain a 0.060 to 0.049 cm ø spot size. A power density of ≈5 kW/cm^2 was obtained at the beginning of laser tests with all of the Ar laser wavelengths and at 1 kW/cm^2 when only 514 nm was used. The transmitted light was monitored with a Coherent Lab Master power meter controller, Coherent LM-10 and LM-45 detectors, and the transmission recorded as a function of time with Lab View software on a PC computer. The transmittance was normalized to the laser output via a beam splitter placed before the focusing lens. Vacuum was achieved with a roughing pump and a liquid nitrogen cold trap. The vacuum level was monitored with a Granville-Phillips convection gauge.

The damage of the optical coating was assumed to be more susceptible to Ar ion wavelength irradiation rather than by the nIR wavelength. One reason was that the coating materials have higher absorptances at shorter wavelengths. Also, all the layers in the coating are designed to be irradiated by and transmit the Ar wavelengths. This increases the probability of exposing any defect sites to the shorter wavelengths. In contrast, only few layer-pairs of the nIR reflector stack bear the brunt of the nIR intensity.

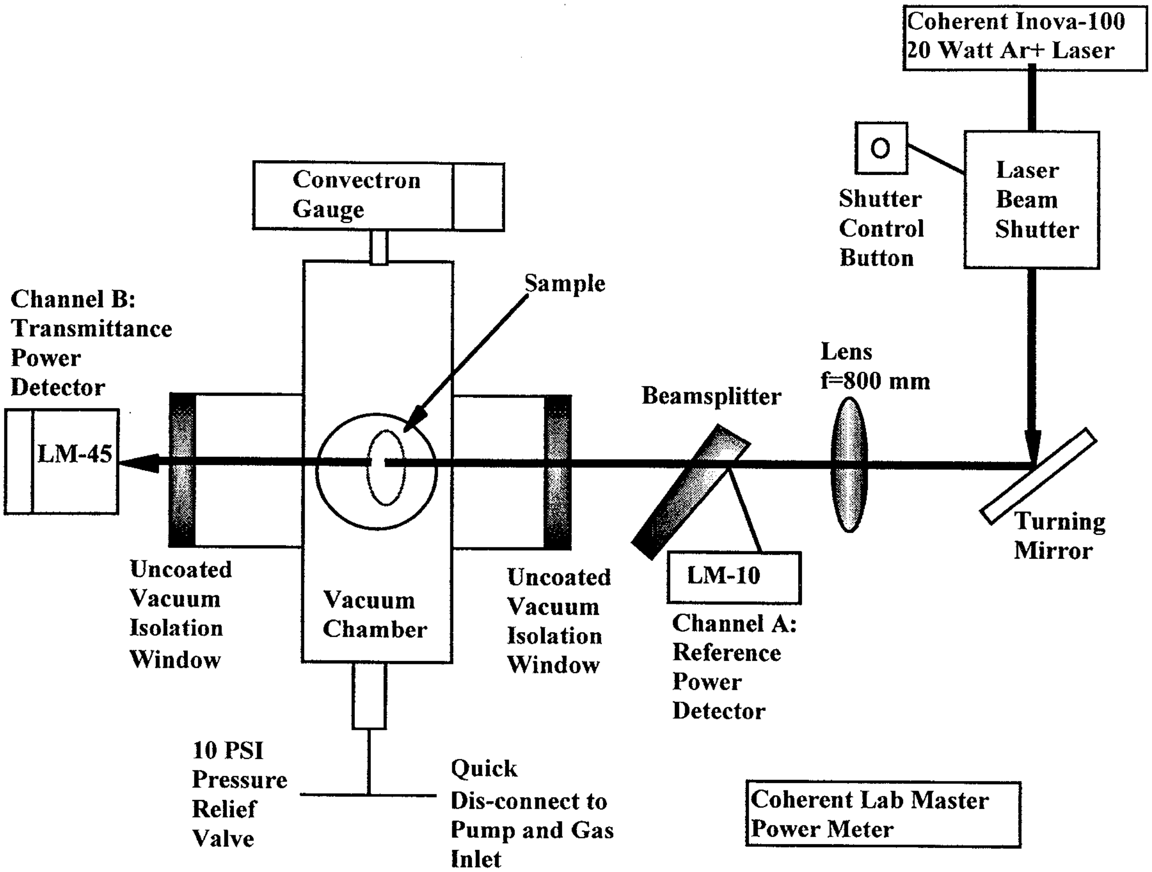


Figure 3 Sketch of the test set-up. The vacuum chamber was 6-way cross made of stainless steel.

Table 1 Ar ion wavelengths and relative power outputs per manufacturer's specification sheet

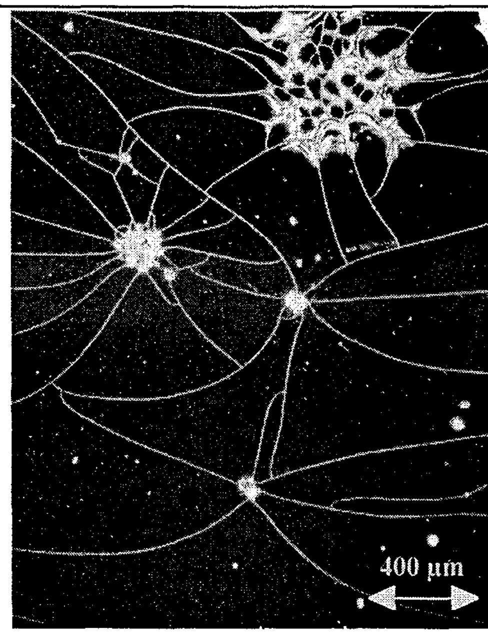
Wavelength (nm)	Relative Power output
514	0.32
501	0.06
496	0.11
488	0.24
476	0.12
472	0.05
465	0.03
457	0.06
454	0.02

3. RESULTS

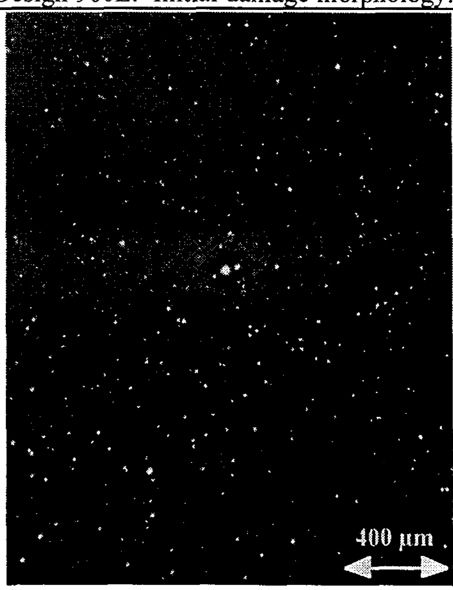
3.1 Damage morphologies

The damage morphologies in Figure 4 were produced during operations and were from the two different designs. Figures 4a and 4b are from Design 900L and Figures 4c and 4d are from Design AAA. Design 900L apparently produced a more tensile stressed coating since the damage morphology in the early (Figure 4a) and later stages (Figure 4b) exhibited craze lines. These craze lines intersect at 90° angles observed in the later stage of damage, indicative of tensile stresses. Design AAA produces a blister type damage morphology which is characteristic of an ejected nodule that had some adhesion to the coating. There was a pit from which the nodule was buried in and collateral damage around the periphery of the pit created by the ejecta pulling away some coating material. [4] These optics were damaged with an illumination of 55 kW/cm^2 of intracavity power density and a pump power density of ≈ 1 kW/cm^2 (2 mm ø spot and 30 W Ar ion laser beam). In comparison, the optics were only exposed to 4 kW/cm^2 from an Ar ion laser in the off-line tests. Other possibilities for different damage morphologies is that different particulates in the cavity contaminated the surfaces prior to damage or that the exposure times were much different for these two optics.

A Design 900L. Initial damage morphology.



B Design 900L Final damage morphology.



C Design AAA damage morphology.

D Design AAA damage morphology. There is a pit at the center, surrounded by a faint circle of debris or coating delamination.

Figure 4 Dichroic coating damage morphologies from two designs, 900L and AAA. Darkfield microscopy was used to view the surface. The smaller points of light are probably caused by coating scatter or particulates

The transmission of both the optics decreased when irradiated with an Ar ion laser beam in an environment of less than 4 mTorr of pressure (Figure 5). The rate of transmission decrease ranged from about 2 to 28 %/day. The wide variation may be dependent on the history of the area tested as to whether it was previously irradiated and/or damaged. When the optics were operated in an oxygen partial pressure of 10 Torr, the transmission increased to nearly the original performance, and degraded at a very slow rate (Figure 6). Design AAA also sustained high transmission performance when irradiated at the lower partial pressure of 1 Torr oxygen and another condition of a partial pressure of air at 10 Torr. Comparing the curves at high and low oxygen partial pressures, the optic took longer to recover at the lower oxygen pressure. The result of operating in a partial pressure of air indicated that constituents of air did not interfere with the oxygenation mechanism that sustains the transmission of the optic.

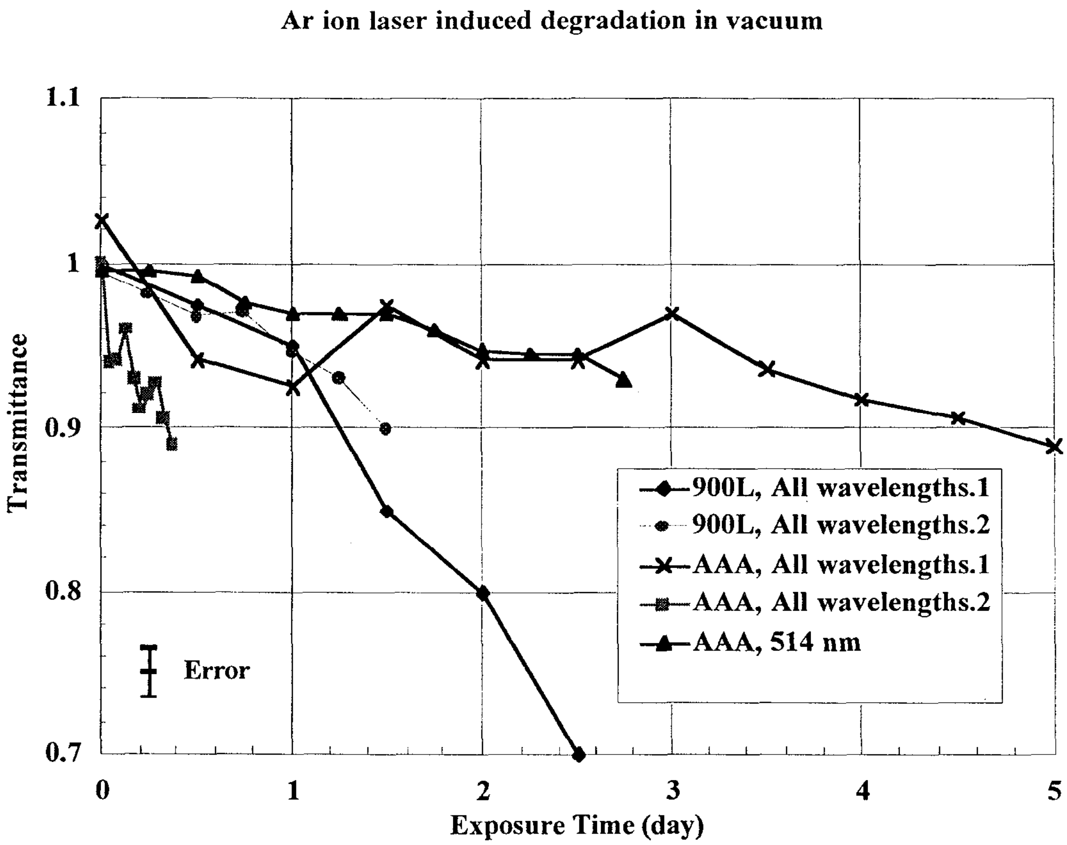


Figure 5 Optic degradation in a vacuum of ≈ 1 mTorr. The test number is noted after the word "wavelengths."

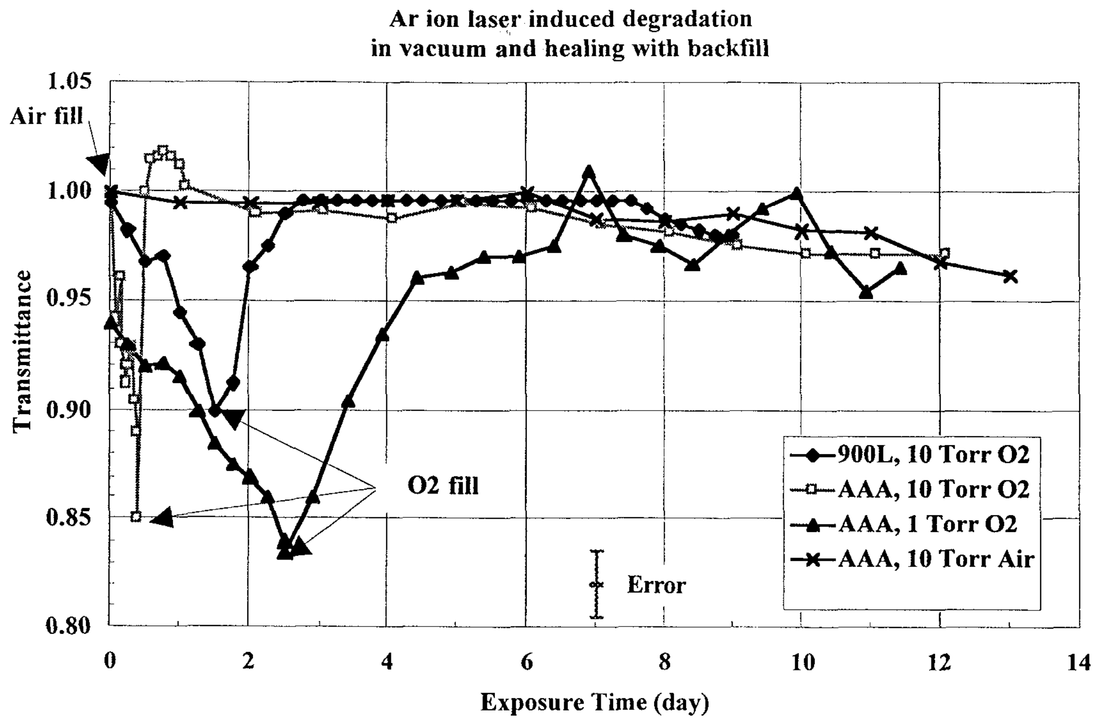


Figure 6 Optical transmission degradation, recovery with light and oxygen, and long term operation in the various oxygen partial pressures noted in the legend.

If the transmission decreased too far, nominally below 80%, the degradation appeared to be irreversible. Intermediate damage was observed with high intensity white-light illumination as changes in the color of the coating. Exposed and unexposed areas of a coating were examined by the electron spectroscopic and chemical analysis technique. No measurable chemical and stoichiometric differences were found between these two areas. Both areas were slightly oxygen deficient after sputter cleaning of the surface. If the degradation continued past 80%, the damage morphologies shown in Figure 4 developed.

A test of a single Ar wavelength, 514 nm, was conducted. The results are plotted in Figure 5, showing that the elimination of the shorter, blue/ultra-violet, wavelengths from the irradiation test did not minimize the transmission degradation rate. This raises the issue of long duration operation of a high average power, visible wavelength, laser system in vacuum.

The transmission recovery appeared to require both the presence of light and oxygen (Figure 7). An optic was tested with oxygen to establish a transmittance baseline. The oxygen was pumped out and the transmittance subsequently begun to decreases. Without breaking vacuum laser propagation was terminated, the test chamber backfilled with 10 Torr of oxygen, and the optic remains in this environment for about 2.5 days. Upon laser illumination, the transmission was observed to be at same value.

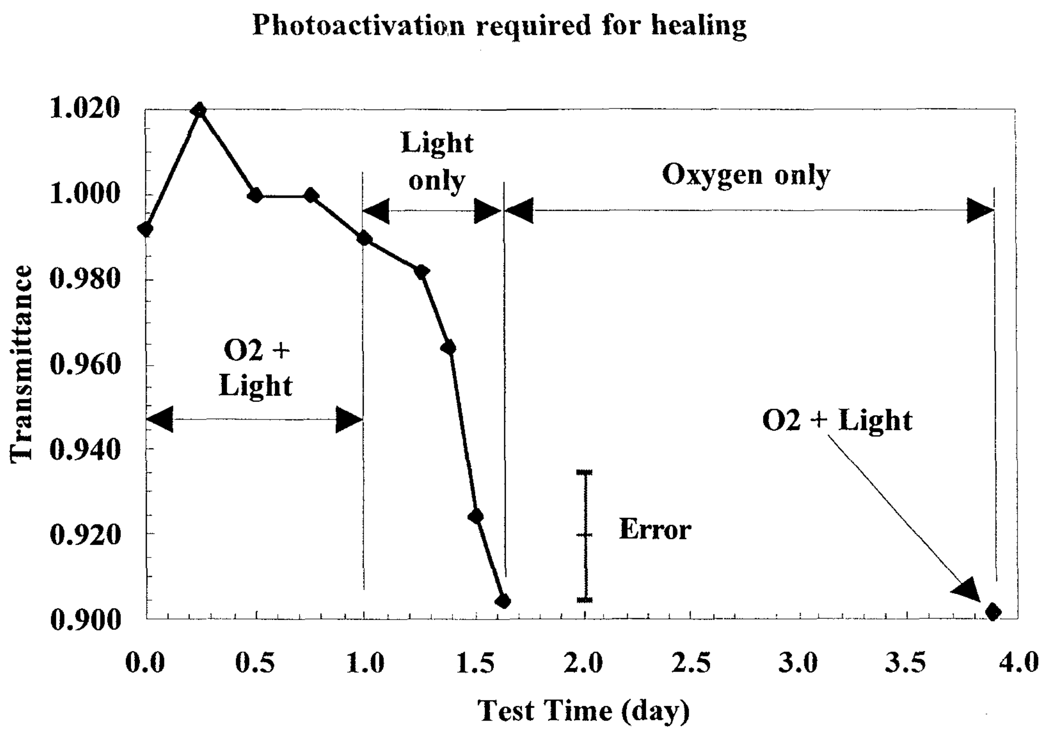


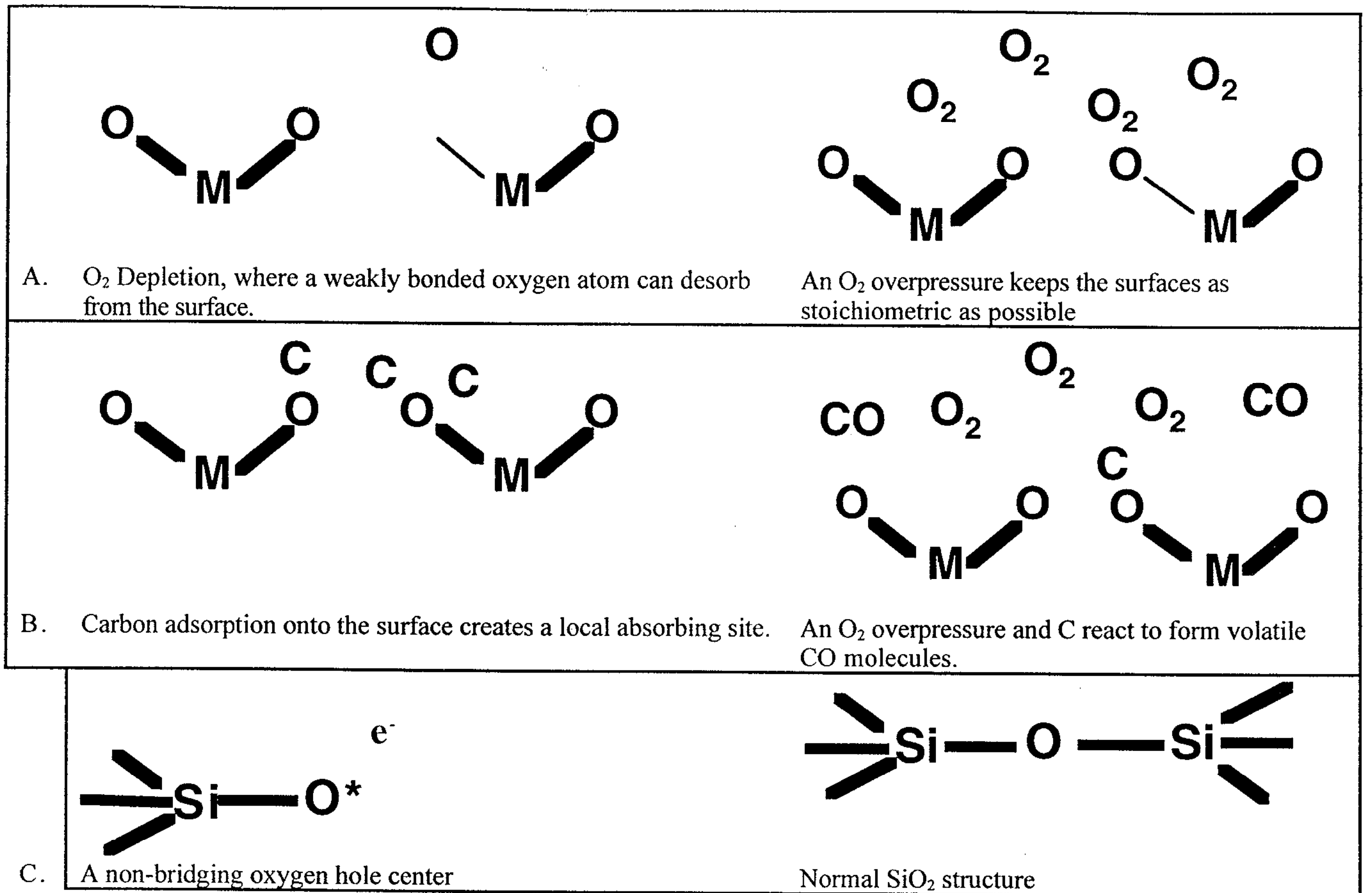
Figure 7 Light and oxygen are both required to heal the optical damage.

4. DISCUSSION

There are several photo-chemistry mechanisms that may cause the transmission degradation. One possible mechanism is photo-induced oxygen depletion from the coating. This mechanism depends on the columnar structure of the e-beam deposited oxides as the transport channels of the oxygen molecules. In the deposition process, refractory metal oxides are e-beam evaporated and deposited onto the substrate in the presence of an oxygen overpressure. The evaporation process can disassociate the oxides into metal and oxygen molecules. The overpressure of oxygen during evaporation increases the oxidation probability of any metallic species deposited at the surface. If the oxidation reactions are not complete, then there may be a high concentration of loosely bonded oxygen atoms to sub-oxides. If there is no overpressure of oxygen during laser irradiation, these oxygen species may be removed during irradiation with high intensity laser light, the metallic-rich area becomes more absorbing, and catastrophic damage eventually occurs. With an oxygen overpressure, the loss of oxygen from the matrix can be quickly replaced with an adjacent molecule from the ambient.

Another proposed mechanism involves photo-chemistry between hydro-carbons and oxygen molecules. The vacuum conditions of the resonator cavity and the test set-up have many sources of hydrocarbons. There are plasticizers from electrical insulation, the rubber o-ring gaskets, and even the pumping system with vacuum oils can contribute molecular hydrocarbons. In the presence of high intensity light, hydrocarbons are disassociated and carbon deposited onto the coating. The carbon increases the surface absorptance and decreases the transmittance of the optic. With an overpressure of oxygen and light, oxygen reacts with the carbon, and forms volatile carbon monoxide which may be pumped away.[5] However, this mechanism does not explain the color change observed on the optic.

A third possibility is similar to ultra-violet induced-degradation, where ultra-violet irradiation is known to create color centers in metal oxides. In fused silica, there is a Non-bridging oxygen hole center which absorbs light at 621 nm and an Al-impurity defect which absorbs at 517 nm.[6] There may be other types of color centers introduced by the deposition process into the coating, which is structurally and chemically more inhomogeneous than bulk material. These other type of color centers may be susceptible to damage by the Ar ion laser wavelengths. However, while this mechanism explains the color change, it does not require the necessity of oxygen for the recovery of the transmission.



A. O_2 Depletion, where a weakly bonded oxygen atom can desorb from the surface. An O_2 overpressure keeps the surfaces as stoichiometric as possible

B. Carbon adsorption onto the surface creates a local absorbing site. An O_2 overpressure and C react to form volatile CO molecules.

C. A non-bridging oxygen hole center. Normal SiO_2 structure

Figure 8 Proposed transmission degradation mechanisms

5. SUMMARY

The summary of the laser irradiation tests under various ambient conditions is in Table 2. High average laser power density Ar ion light illuminating a dichroic in vacuum caused the transmission to decrease. Other than the different damage morphologies, dichroics of two different designs perform identically. They degraded when irradiated in vacuum with a high average power laser density from an Ar ion laser, and their performance recovered to near-normal transmission levels when irradiated in oxygen partial pressures greater than 1 Torr. The presence of other atmospheric gases did not appear to interfere with the high transmission performance. If the transmission degradation was not too severe, the performance of the optic was recovered by illuminating in the overpressure of oxygen. However, exposure to oxygen alone was not sufficient to recover the optical transmission performance.

Table 2 Summary of the nIR dichroic performance with vacuum and wavelengths.

Pressure (Torr)	Gas	Ar ion wavelengths	Transmittance
< 0.001	Air	All	Degradation
1	Oxygen	All	Steady
10	Oxygen	All	Steady
30	Oxygen	All	Steady
10	Air	All	Steady
< 0.004	Air	514 nm	Degradation

A work-around solution to the transmission degradation was to operate the resonator in a partial pressure of oxygen. Further work is needed to determine the relationship between power density and the level of oxygen required to sustain the optical performance. Another idea that addresses the oxygen depletion mechanism is to test a dichroic made of fluorides instead of all oxides. However an all-fluorides multi-layer would be difficult to make because the layers are more stressful than oxide layers. [7,8] The main thrust then may be to use fluorides on the top-most layers which would act as an oxygen diffusion barrier. To make the experiment even simpler, anti-reflective instead of dichroic coatings made of metal oxides and metal fluorides for the Ar wavelengths can be tested and compared. If the oxygen overpressure only works with the oxide AR. this would support the oxygen depletion mechanism. If the oxygen overpressure works in both material cases, this would support the hydrocarbon mechanism.

Another approach is to perform mass spectroscopy during the laser irradiation reaction to verify and quantify evolution of volatile species. Observation of carbon monoxide or carbon dioxide species during irradiation with an oxygen overpressure would support the hydro-carbon mechanism. Observation of increased oxygen levels during vacuum irradiation would supports the oxygen depletion mechanism.

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